

THERMODYNAMICS OF HETEROGENEOUS AND ANISOTROPIC NONLINEAR FERROELASTIC CRYSTALS

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ABSTRACT. In a previous paper, in a geometrized framework for the description of simple materials with internal variables, the specific example of ferroelastic crystals with anisotropy grain-tensors *à la* Maruszewski was considered and the relevant structure of the entropy 1-form was derived. In this contribution the linear morphism defined on the fibre bundle of the process and the transformation induced by the process are obtained as new results within the geometrical model. Furthermore, Clausius-Duhem inequality for these media is exploited, and, using a Maugin technique (see also Coleman-Noll procedure), the laws of state, the extra entropy flux and the residual dissipation inequality are worked out. Finally, following Maugin, the heat equation in the first and the second form is derived.

1. Introduction

In a previous paper [1] a geometrization technique for thermodynamics of simple materials with internal variables [2]-[8] was applied to the specific example of ferroelastic crystals (see [9]-[13]) and the expressions for the existence of an entropy function and the entropy 1-form were derived. The model of ferroelastic crystals developed in [9]-[13] is based on a concept of *anisotropy grain structure* first introduced in [14]. The idea used consists in the assumption that the body as a whole is homogeneous and isotropic, where there exist particular physical fields responsible for its internal structure and geometry, described by internal variables [14]. In this way the balance equations for the mass, the momentum, the moment of momentum and the internal energy, and the rate equation for the heat flux vector are supplemented by the evolution equations for the anisotropy-grain tensors forming altogether a coupled system of equations describing the behavior of the ferroelastic body considered. The proper form of these equations has been derived from the thermodynamical models presented in [9]-[13]. The stress wave propagation and some explicit examples of stress tensors in these models were considered in [13] and [10]-[12]. In this contribution the linear morphism defined on the fibre bundle of the process and the transformation induced by the process are obtained as new results within the geometrical model developed in [1] for the description of ferroelastic crystals (see also [15] and [16] for the geometrization technique applied). Furthermore, we exploit the Clausius-Duhem inequality for these media and, using a Maugin technique [17] (see also Coleman-Noll procedure in [18]), we derive the extra entropy flux, the residual dissipation inequality and the laws of state, where as relevant result the contributions in the total stress tensor expressed in terms of internal variables are rewritten in terms of macroscopic variables (the

Duhamel-Neumann tensor stress and the nonlinear strain tensor). Finally, the heat equation in the first and the second form is obtained following Maugin [17].

2. The anisotropy-grain tensor model

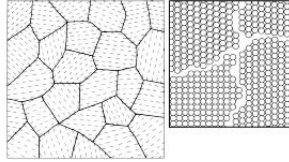


FIGURE 1. Internal structure of the body. Anisotropic character of grains (after [19])

Now, we recall the model developed by Maruszewski in [14] for heterogeneous and/or anisotropic media. The ferroelastic crystal can be defined as a crystal which has at least two orientation states that differ from each other with respect to at least one of the components of the spontaneous strain tensor. A suitably oriented uniaxial stress can often switch one ferroelastic orientation state to another so that the ferroelastic domains can be made to grow or shrink by applying such a driving force in complete analogy with domains in ferroelectric and ferromagnetic materials. In [14] it was assumed that a heterogeneous body \mathcal{B} is formed by several kinds of grains with different anisotropies (see Fig.1 after [19]). The families of different species of grains are labeled by an index α (α running from 1 to an integer r , the total number of species) and the families of different anisotropies occurring in the body are labeled by a further index β (β running from 1 to s , the total number of *anisotropies*). A third index γ is introduced to distinguish between line anisotropies ($\gamma = 1$) and planar anisotropies ($\gamma = 2$). Now, following Maruszewski, the body as a whole is regarded as homogeneous and isotropic in which particular physical fields responsible for its internal structure and geometry exist. Such fields are modeled by so-called internal variables, having tensorial character and defined by the *Anisotropy-Grain tensor* (or shortly, the AG-tensor)

$$(1) \quad \mathbf{I}^{\alpha\beta}(t, \mathbf{x}) = V^\alpha n^{\alpha\beta}(t, \mathbf{x}) \mathbf{A}^{\beta\gamma}, \quad (\alpha = 1, 2, \dots, r; \beta = 1, 2, \dots, s; \gamma = 1, 2),$$

where there is no summation over α and β . In equation (1) V^α denotes the characteristic dimension of the α -grain or domain field possessing β anisotropy (in the 3D case it is the characteristic volume of the α -grain, in the 2D case the characteristic surface S^α of the α -grain, in the 1D case the characteristic length L^α (diameter) of the α -grain). Finally, $n^{\alpha\beta}(t, \mathbf{x})$ is the *number density* of the α -grain with anisotropy β defined as follows

$$(2) \quad 3D : n^{\alpha\beta}(t, \mathbf{x}) = \frac{dN^{\alpha\beta}(t, \mathbf{x})}{dV^\alpha}, \quad 2D : n^{\alpha\beta}(t, \mathbf{x}) = \frac{dN^{\alpha\beta}(t, \mathbf{x})}{dS^\alpha}, \quad 1D : n^{\alpha\beta}(t, \mathbf{x}) = \frac{dN^{\alpha\beta}(t, \mathbf{x})}{dL^\alpha},$$

where $N^{\alpha\beta}(t, \mathbf{x})$ is the corresponding total number of grains with β anisotropies.

Moreover, in equation (1) the tensor $\mathbf{A}^{\beta\gamma}$ reflects the anisotropy (geometry) of a grain and it is defined as follows (see Fig.2): $\mathbf{A}^{\beta 1} = \mathbf{t}^\beta \otimes \mathbf{n}$, $\mathbf{A}^{\beta 2} = \mathbf{n}^\beta \otimes \mathbf{n}$, for line anisotropies and for planar anisotropies, respectively, with \mathbf{n} the unit normal vector to the cross section $\delta\mathcal{B}$ of the domain $d\mathcal{B}$ of the body, \mathbf{t}^β the tangent unit vector to the line anisotropic structure β and \mathbf{n}^β the

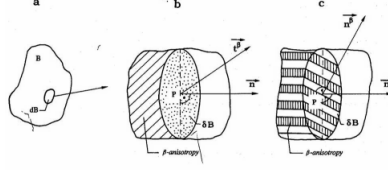


FIGURE 2. Element $d\mathcal{B}$ of a domain in the case of line (b) and planar (c) anisotropies. $\delta\mathcal{B}$ is the cross-section of $d\mathcal{B}$ (after [14])

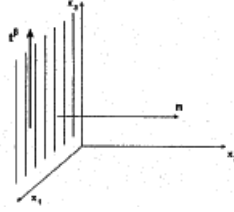


FIGURE 3. Line anisotropy model (after [13])

unit normal vector to the surface anisotropy. $\Gamma^{\alpha\beta}$ may be regarded as a sort of order parameter for ferroelasticity, in the sense of Landau [20], provided one assumes that anisotropies disappear in the ferroelastic crystal when θ reaches the critical value θ_c (i.e., $\mathbf{A}^{\beta\gamma} = 0$ for $\theta \geq \theta_c$). A few specific examples have been analyzed in detail in [9], [12] and in [13]. Let us consider now a case of *line anisotropy of the body* spontaneously deformed (see Fig. 3 after [13]). We assume that all the lines or the fibers (infinite quantity because of the continuous model) are placed in the plane x_1x_2 and parallel to x_3 -axis. Since we consider a single-domain (one grain) medium, the anisotropy-grain tensor reads

$$(3) \quad \Gamma_{ij}^{\beta} = \Gamma_{ij} = A_{ij}^{\beta 1}(\mathbf{x}, t) = \mathbf{t}^{\beta} \otimes \mathbf{n}, \quad \mathbf{t}^{\beta} = \{0, 0, 1\}, \quad \mathbf{n} = \{0, 1, 0\}.$$

Hence, only $\Gamma_{32} \neq 0$ since

$$(4) \quad \mathbf{A}^{\beta 1} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}.$$

Let us consider now a case of *plane anisotropy of the body* spontaneously deformed (see Fig. 4 after [13]). We assume that all the planes (infinite quantity because of the continuous model) are parallel to the plane x_1x_3 . Since we consider a single grain medium, the anisotropy-grain tensor reads

$$(5) \quad \Gamma_{ij}^{\beta} = \Gamma_{ij} = A_{ij}^{\beta 2}(\mathbf{x}, t) = \mathbf{n}^{\beta} \otimes \mathbf{n}, \quad \mathbf{n}^{\beta} = \{0, 1, 0\}, \quad \mathbf{n} = \{0, 1, 0\}.$$

Hence, only $\Gamma_{22} \neq 0$ since

$$(6) \quad \mathbf{A}^{\beta 2} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

In the case of an *anisotropic layered structure of a grain*, the total anisotropy of a grain is described

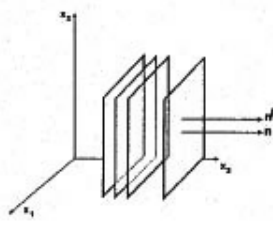


FIGURE 4. Plane anisotropy model (after [13])

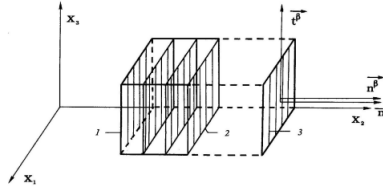


FIGURE 5. Anisotropic layered structure (schematic draw): (1) grain, (2) planar anisotropy, (3) line anisotropy (after [14])

by the superposition of the line and planar anisotropies (see [14]). Hence, we can introduce the total anisotropy tensor as $A_{ij}^\beta = A_{ij}^{\beta 1} + A_{ij}^{\beta 2}$. A simple example is presented on Fig. 5 (see [14]), where

$$\mathbf{t}^\beta = \{0, 0, 1\}, \quad \mathbf{n} = \{0, 1, 0\}, \quad \mathbf{n}^\beta = \{0, 1, 0\}, \quad \mathbf{A}^{\beta 1} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}, \quad \mathbf{A}^{\beta 2} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

Hence, the total anisotropy tensor reads

$$(7) \quad A_{ij}^\beta = A_{ij}^{\beta 1} + A_{ij}^{\beta 2} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 1 & 0 \end{pmatrix}.$$

It shows that it is possible to describe quite complex structures. In the limiting case of *homogeneous body*, when the body does not consist of grains we have (3D) [14]

$$(8) \quad V^{\alpha\beta} = V, \quad n^{\alpha\beta}(t, \mathbf{x}) = \frac{N^{\alpha\beta}(t, \mathbf{x})}{V}, \quad N^{\alpha\beta}(t, \mathbf{x}) = 1, \quad \Gamma^{\alpha\beta}(t, \mathbf{x}) = \mathbf{\Gamma}^\beta = \mathbf{A}^{\beta\gamma}.$$

It means that in such a case the tensor $\Gamma^{\alpha\beta}$ describes, as internal variable, only the anisotropy.

In the other limiting case of the *isotropic body* the tensor $\mathbf{A}^{\beta\gamma}$ takes the following form:

$\mathbf{A}^{\beta\gamma} = \mathbf{I}$, hence $\mathbf{\Gamma}^\alpha = V^\alpha n^{\alpha\beta} \mathbf{I}$ describing only the grain structure of the body.

The above defined anisotropy-grain tensor $\mathbf{\Gamma}^{\alpha\beta}$ can describe heterogeneous and anisotropic media as an internal variable in many situations. Evolution of grain structure and anisotropy of materials occurs during many processes accompanied by mechanical deformation and thermal treatments. Such phenomena as: recrystallization, phase transitions, twinning, grain growth and similar phenomena, are typical during annealing, cooling, cold-working or plastic deformations [14]. The models of ferroelastic bodies may have relevance in the

study of concrete materials such as, e.g., steel, martensitic plates in metals and others. On Fig. 6 the evolution of structure and anisotropy in materials is shown (after [21]).

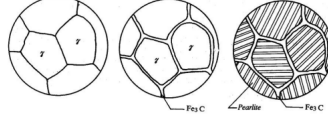


FIGURE 6. Evolution of a microstructure of steel during cooling (after [21])

3. Fundamental laws

In [10], in the framework of the extended rational thermodynamics with internal variables, using the standard Cartesian tensor notation in a rectangular coordinate system, a model was developed for a heterogeneous and anisotropic non linear thermoelastic body, in which the following fields interact with each other: *the elastic field* described in the case of finite deformations by the total stress tensor t_{ij} and the nonlinear Euler strain tensor E_{ij} ; *the thermal field* described by the temperature θ and the heat flux q_i ; *the grain-anisotropy field* described by the internal variable Γ_{ij} , in the case that there exists one type (size) of grain ($\alpha = 1$) and one kind of anisotropy ($\beta = 1$). Thus, the independent variables are represented by the set

$$(9) \quad C = \{E_{ij}, \theta, \theta_{,i}, q_i, \Gamma_{ij}\}.$$

The specific choice shows that the relaxation properties of the thermal field (second sound) are taken into account in agreement with the general philosophy of the extended thermodynamics. The corresponding effects for the mechanical field and for the anisotropy-grain field are ignored, i.e. viscous properties of the body are excluded from the consideration and the field Γ_{ij} is not of kinetic character. All the processes occurring in the body considered are governed by two groups of laws. The first one deals with the balance laws:

the continuity equation

$$(10) \quad \dot{\mu} + \mu v_{i,i} = 0,$$

where μ is the mass density and $\mathbf{v} = \dot{\mathbf{u}}$ (being $\mathbf{u} = (u_i)$ the displacement vector and the superimposed dot denotes the material derivative);

the momentum balance

$$(11) \quad \mu \dot{\mathbf{v}} - \text{div} \mathbf{t} - \mathbf{f} = 0,$$

where \mathbf{f} denotes the body force and \mathbf{t} is the total stress tensor given by

$$(12) \quad \mathbf{t} = \mathbf{T} - \boldsymbol{\sigma}^{TE} - \boldsymbol{\sigma}^{HA}$$

(see equation (2.19) of reference [10] for the law of state (12) defining the *total stress tensor* \mathbf{t}). First, we mention that the model developed in [10] refers to large deformations, so that $\mathbf{T} = \|T_{ij}\|$ is here the symmetric Duhamel-Neumann stress tensor defined as

$$(13) \quad T_{ij} = \mu \frac{\partial \Psi}{\partial E_{ij}}.$$

The extra terms in (12) account for the grain-anisotropy. The term σ^{TE} corresponds to the *thermoelastic stress* and in components $\|\sigma_{ij}^{TE}\|$ is given by

$$(14) \quad \sigma_{ij}^{TE} = \Pi_s^q q_j E_{si} + \Pi_i^q q_k E_{jk},$$

where $\mathbf{q} = (q_i)$ is the *heat flux vector*, $\Pi^q = (\Pi_i^q)$ is the *affinity* defined by $\Pi_i^q = \mu \frac{\partial \Psi}{\partial q_i}$, and $\Psi = e - \theta S$, e and S are the *Helmholtz free energy*, the *internal energy* and the *entropy* per unit mass, respectively. In [10] it was derived that Ψ depends on the following set of variables $\Psi = \Psi(\mathbf{E}, \theta, \mathbf{q}, \Gamma)$. The non linear Eulerian strain tensor E_{ij} is a symmetric rank-2 tensor defined by $E_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i} - u_{m,i}u_{m,j})$. It describes the *elastic field* in the case of finite deformations. In the general case (α kinds of grains and β anisotropies), the second extra term of (12) accounts for the interaction of the grain-anisotropy field with the mechanical field. It is defined by

$$\sigma_{ij}^{HA} = \sum_{\alpha, \beta} \sigma_{ij}^{HA\alpha\beta} \text{ with } \sigma_{ij}^{HA\alpha\beta} = \eta_{rs}^{\alpha\beta} \Gamma_{js}^{\alpha\beta} E_{ri} + \eta_{rs}^{\alpha\beta} \Gamma_{rj}^{\alpha\beta} E_{si} + \eta_{ri}^{\alpha\beta} \Gamma_{rs}^{\alpha\beta} E_{js}$$

and $\eta_{ij}^{\alpha\beta} = \mu \frac{\partial \Psi}{\partial \Gamma_{ij}^{\alpha\beta}}$ an affinity;

the moment of momentum balance $\varepsilon_{ijk} t_{jk} = 0$,

which results from the assumption that the medium has no spin, microrotational or other skew-symmetric features, it means that the total stress tensor is symmetric (see equations (3.3)-(3.6) in [10], where, using the laws of state, a constitutive relation was worked out for a symmetric total stress tensor according to particular assumptions);

the internal energy balance

$$(15) \quad \mu \dot{e} - p_{(i)} + \text{div} \mathbf{q} - \mu r = 0,$$

with r the heat radiation per unit of mass and $p_{(i)}$ the power of internal forces defined by

$$(16) \quad p_{(i)} = \mathbf{t} \cdot \mathbf{L} = (\mathbf{T} - \sigma^{TE} - \sigma^{HA}) \cdot \mathbf{L},$$

where $\mathbf{L} = \nabla \mathbf{v}$ is the gradient of the velocity. In the following sections we use the relation $\mathbf{L} = \dot{\mathbf{F}}\mathbf{F}^{-1}$ expressing \mathbf{L} in terms of the gradient of the deformation \mathbf{F} .

A second group of laws deals with the rate properties of the heat flux and the anisotropy-grain field

$$(17) \quad \dot{\mathbf{q}}^* = \mathbf{Q}(C), \quad \dot{\Gamma}^* = \mathbf{G}(C),$$

where $\dot{q}_i^* = \dot{q}_i - \Omega_{ij} q_j$, $\dot{\Gamma}_{ij}^* = \dot{\Gamma}_{ij} - \Omega_{ik} \Gamma_{kj} + \Gamma_{ik} \Omega_{kj}$, $\Omega_{ij} = \frac{1}{2}(v_{i,j} - v_{j,i})$ is the antisymmetric part of $L_{ij} = v_{i,j}$ and the superimposed asterisk indicates the Zaremba-Jaumann time derivative (see [22]). To be sure that the physical processes occurring in the body considered are real, all the admissible solutions of the proposed evolution equations have to satisfy the following *entropy inequality*

$$(18) \quad \mu \dot{S} + \nabla \cdot \mathbf{J}_S \geq 0, \quad \text{where} \quad \mathbf{J}_S = \frac{1}{\theta} \mathbf{q} + \mathbf{k},$$

with \mathbf{J}_S the entropy flux and \mathbf{k} an additional term called extra entropy flux density (see [17] and [23]-[26]). The set of the constitutive functions are

$$(19) \quad \mathbf{Z} = \{\mathbf{t}, e, S, \mathbf{k}, \mathbf{Q}, \mathbf{G}\}.$$

In [9]-[13] constitutive equations $\mathbf{Z} = \tilde{\mathbf{Z}}(C)$ were obtained for ferroelastic crystals in different cases. The entropy inequality (18) was analyzed by Liu's theorem [27], and some constitutive relations were obtained, using Smith theorem [28], with the help of isotropic polynomial representations of proper constitutive functions satisfying the objectivity and material frame indifference principles (see [29]-[31]). In [10] the constitutive relation (12) for the total stress tensor was obtained expanding the free energy in Taylor series with respect to a particular natural state. In [12] the most general expression for \mathbf{G} in the case of one-domain ferroelastic crystals (i.e., $r = s = 1$) was explicitly calculated (see equ. (3.24)) in the following form: $\tau^\Gamma \dot{\Gamma}_{ij} = a\delta_{ij} + bt_{ij} + c\Gamma_{ij} + N(\mathbf{q}, \nabla\theta, \mathbf{t}, \Gamma)$, where τ^Γ denotes the relaxation time for the spontaneous strain field, the coefficients a , b , c depend on the temperature θ and the set of invariants built on \mathbf{t} , $\nabla\theta$, Γ , \mathbf{q} (see (3.18), (3.22) and (3.25)), while the non-linear part N depends also on \mathbf{t} , $\nabla\theta$, Γ , \mathbf{q} and can be expanded into a polynomial expression with coefficients depending on θ and the same invariants. The linearized theory ($N = 0$, $a = 0$) was considered explicitly in [13].

4. A geometric model for ferroelastic crystals

Now, we present a short review of a geometric model for ferroelastic crystals elaborated in [1], where following [2]-[4], introducing the concepts of *process* and *transformation*, we have derived the expressions for the existence of an entropy function and the entropy 1-form. We have considered a material point and we have defined the state space at time t as the set B_t of all state variables which "fit" the configuration of the element at time t . B_t is assumed to have the structure of a finite dimensional manifold. The "total state space" is the disjoint union $\mathcal{B} = \bigcup_t \{t\} \times B_t$ with a given natural structure of fibre bundle over \mathbb{R} , where time flows [2]-[4]. If the instantaneous state space B_t does not vary in time (i.e. there is an abstract space B such that $B_t \simeq B$ for all instant of time t), then the total state space \mathcal{B} has the topology of the Cartesian product $\mathcal{B} \simeq \mathbb{R} \times B$ (see [2]-[4]). Moreover, we consider an abstract space of *processes* [2]-[6], i.e. a set Π of functions $P_t^i : [0, t] \rightarrow \mathcal{G}$, where $[0, t]$ is any time interval, the space \mathcal{G} being a suitable target space defined by the problem under consideration, i a label ranging in an unspecified index set for all allowed processes and $t \in \mathbb{R}$ the so called *duration* of the process. Then, a continuous function is defined (see [2]-[8])

$$(20) \quad \chi : (t, P_t^i) \in \mathbb{R} \times \Pi \rightarrow \rho_t^i \in C^0(B, B),$$

with $\rho_t^i : b \in D_t^i \subseteq B \rightarrow \rho_t^i(b) = b_t \in R_t^i \subseteq B$, so that for any instant of time t and for any process $P_t^i \in \Pi$ a continuous mapping, ρ_t^i , called *transformation induced by the process* is generated, which gives point by point a correspondence between the initial state b and the final state $\rho_t^i(b) = b_t$. Now, we introduce a function of time

$$(21) \quad \lambda_b^i(\tau) = \begin{cases} b & \text{if } \tau = 0 \\ \rho_t^i(b) & \text{if } \tau \in]0, t] \end{cases} \quad \text{with } b \in D_t^i$$

such that the transformation for the medium is given by $\delta : \mathbb{R} \longrightarrow \mathbb{R} \times B$

$$(22) \quad \delta : \tau \in \mathbb{R} \longrightarrow \delta(\tau) = (\tau, \lambda_b^i(\tau)) \in \mathbb{R} \times B.$$

With these positions the transformation is interpreted as a curve δ in the union of all state spaces such that it intersects the instantaneous state space just once. In [1], taking into account the nonconventional thermodynamical model developed in section 3 of this paper,

it was assumed that the state variables are the *deformation gradient* \mathbf{F} , the *internal energy* e (by a Legendre transformation the temperature θ was replaced with the internal energy e), the vector $\beta = \nabla\theta$, the heat flux vector \mathbf{q} and the Anisotropy-Grain tensor $\mathbf{\Gamma}$ in the case that one type (size) of grain ($\alpha = 1$) and one kind of anisotropy ($\beta = 1$). Then, the full state space is

$$(23) \quad \mathcal{B} = \text{Lin}(\mathcal{V}) \oplus \mathbb{R} \oplus \mathcal{V} \oplus \mathcal{V} \oplus W,$$

where W is a vector space accounting for the internal variable $\mathbf{\Gamma}$. In the general case of α grains ($\alpha = 1, 2, \dots, r$) and β anisotropies ($\beta = 1, 2, \dots, s$), $W = \otimes_{\alpha, \beta} T_2^0(\mathcal{V})$ is given by $r.s$ copies of $T_2^0(\mathcal{V})$, $\mathcal{V} \simeq \mathbb{R}^3$, each one corresponding to a grain specie α with anisotropy β , being $T_2^0(\mathcal{V})$ a rank-2 tensor. Accordingly our state variables are $(\mathbf{F}, e, \nabla\theta, \mathbf{q}, \mathbf{\Gamma})$ and the process P_t^i is described by the following functions

$$P_t^i = [\mathbf{L}(\tau), h(\tau), \gamma(\tau), \mathbf{Q}(\tau), \mathbf{\Lambda}(\tau)], \quad \text{where} \quad h(\tau) = -\nabla \cdot \mathbf{q}, \quad \gamma(\tau) = \dot{\beta}.$$

The space \mathcal{G} is given by $\mathcal{G} = \text{Lin}(\mathcal{V}) \oplus \mathbb{R} \oplus \mathcal{V} \oplus \mathcal{V} \oplus W$. Moreover, the constitutive functions θ , \mathbf{t} , \mathbf{Q} and $\mathbf{\Lambda}$ are defined in the following way

$$\theta : \mathbb{R} \times B \longrightarrow \mathbb{R}^{++} \quad \mathbf{t} : \mathbb{R} \times B \longrightarrow \text{Sym}(\mathcal{V})$$

$$\mathbf{Q} : \mathbb{R} \times B \longrightarrow \mathcal{V} \quad \mathbf{\Lambda} : \mathbb{R} \times B \longrightarrow W,$$

where \mathbb{R}^{++} is the set of real positive numbers, \mathcal{V} and W are vector spaces accounting for \mathbf{Q} and $\mathbf{\Lambda}$, respectively.

In this paper we assume that for each pair (P_t^i, b) , the following dynamical system holds

$$(24) \quad \begin{cases} \dot{\mathbf{F}} = \mathbf{L}(\tau)\mathbf{F}(\tau) \\ \mu\dot{e} = \mathbf{t}(\delta) \cdot \mathbf{L}(\tau) + h(\tau) \\ \dot{\beta} = \gamma(\tau) \\ \dot{\mathbf{q}} = \mathbf{Q}(\delta) \\ \dot{\mathbf{\Gamma}} = \mathbf{\Lambda}(\delta), \end{cases}$$

differing from that one presented in the previous paper [1], because in the rate equations of the system (24) for the heat flux and the anisotropy-grain field we have taken into consideration the material derivatives of these quantities and not the Zaremba-Jaumann time derivatives as in (17) (see [29]-[31] for this choice). Equation $(24)_2$ is given by (15), when the heat radiation per unit of mass is disregarded.

In this paper, as new results, we derive the linear morphism G , defined on the fibre bundle of the process determined by the system (24), and the transformation induced by the process $\rho_t^i(b)$. The linear morphism G , defined in the following way

$$G : B \times \mathcal{G} \simeq TB \longrightarrow TB,$$

has the following form

$$(25) \quad G : (\mathbf{F}, e, \beta, \mathbf{q}, \mathbf{\Gamma}, \mathbf{L}, h, \gamma, \mathbf{Q}, \mathbf{\Lambda}) \rightarrow (\mathbf{F}, e, \beta, \mathbf{q}, \mathbf{\Gamma}, \dot{\mathbf{F}}, \dot{e}, \dot{\beta}, \dot{\mathbf{q}}, \dot{\mathbf{\Gamma}}),$$

which in matrix form is expressed by:

$$\begin{pmatrix} \mathbf{F}, e, \beta, \mathbf{q}, \mathbf{\Gamma}, \dot{\mathbf{F}}, \dot{e}, \dot{\beta}, \dot{\mathbf{q}}, \dot{\mathbf{\Gamma}} \end{pmatrix}^T = \begin{pmatrix} \mathbf{I} & \mathbf{0} \\ \mathbf{0} & \mathbf{A} \end{pmatrix} (\mathbf{F}, e, \beta, \mathbf{q}, \mathbf{\Gamma}, \mathbf{L}, h, \gamma, \mathbf{Q}, \mathbf{\Lambda})^T,$$

with

$$(26) \quad \mathbf{A} = \begin{pmatrix} \mathbf{F} & 0 & 0 & 0 & 0 \\ \frac{\mathbf{t}}{\mu} & \frac{1}{\mu} & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}.$$

The transformation $\rho_t^i(b)$ is given by (see [4]):

$$(27) \quad \rho_t^i(b) = \begin{cases} \mathbf{F} &= \int_0^t [\mathbf{L}(\tau)\mathbf{F}(\tau)] dt \\ e &= \int_0^t \frac{1}{\mu} [\mathbf{t}(\delta) \cdot \mathbf{L}(\tau) + h(\tau)] dt \\ \beta &= \int_0^t [\gamma(\tau)] dt \\ \mathbf{q} &= \int_0^t [\mathbf{Q}(\delta)] dt \\ \Gamma &= \int_0^t [\Lambda(\delta)] dt. \end{cases}$$

In this geometrical structure, following standard procedures (see [2]-[4] and [8]), in [1] an "entropy function" was introduced, related to a transformation between the initial state b and the state $\rho_t^i(b) = b_t$, by setting:

$$(28) \quad s(\rho_t^i, b, t) = - \int_0^t \frac{1}{\mu} \nabla \cdot \mathbf{J}_s d\tau = \int_0^t -\frac{1}{\mu\theta} \nabla \cdot \mathbf{q} d\tau + \int_0^t \frac{1}{\mu\theta^2} \mathbf{q} \cdot \nabla \theta d\tau - \int_0^t \frac{1}{\mu} \nabla \cdot \mathbf{k} d\tau,$$

which in turn defines a 1-form Ω in $\mathbb{R} \times B$ called the *entropy 1-form*. Using (24)₂ we have $-\nabla \cdot \mathbf{q} = \mu \dot{e} - \mathbf{t} \cdot (\dot{\mathbf{F}}\mathbf{F}^{-1})$. So that the final expression for $s(\rho_t^i, b, t)$ is

$$(29) \quad s(\rho_t^i, b, t) = \int_\delta \Omega \text{ with } \Omega = \frac{1}{\theta} \left[-\frac{1}{\mu} \mathbf{t}\mathbf{F}^{-T} \cdot d\mathbf{F} + de + \frac{1}{\mu} \left(\frac{1}{\theta} \mathbf{q} \cdot \nabla \theta - \theta \nabla \cdot \mathbf{k} \right) \right] d\tau,$$

where the algebraic rule $\mathbf{t} \cdot (\dot{\mathbf{F}}\mathbf{F}^{-1}) = (\mathbf{t}\mathbf{F}^{-T}) \cdot \dot{\mathbf{F}}$ has been used, with $\mathbf{F}^{-T} = (\mathbf{F}^{-1})^T$ (T being matrix transposition). Expressions of \mathbf{t} , \mathbf{q} and \mathbf{k} were derived in [10] (see also [9], [11]-[13] in which ferroelastic crystals were studied in different cases). Thus, the entropy function is now calculated as an integral along a path into the space $\mathbb{R} \times B$ of all thermodynamic variables together with the independent time variable. In components the entropy 1-form Ω becomes:

$$(30) \quad \Omega = \omega_\mu dq^\mu + \omega_0 dt = \omega_A dq^A, \quad (A = 1, 2, \dots, 6),$$

where

$$q^A = (\mathbf{F}, e, \nabla \theta, \mathbf{q}, \Gamma, t)$$

and

$$\omega_A = \left(-\frac{1}{\mu\theta} \mathbf{t}\mathbf{F}^{-T}, \frac{1}{\theta}, 0, 0, 0, \frac{1}{\mu\theta^2} \mathbf{q} \cdot \nabla \theta - \frac{1}{\mu} \nabla \cdot \mathbf{k} \right).$$

Thus, by differentiation a 2-form is derived:

$$(31) \quad d\Omega = \frac{1}{2} A_{\mu\lambda} dq^\mu \wedge dq^\lambda + E_\lambda dt \wedge dq^\lambda, \text{ where } A_{\mu\lambda} = \partial_\mu \omega_\lambda - \partial_\lambda \omega_\mu; \text{ and } E_\lambda = \partial_0 \omega_\lambda - \partial_\lambda \omega_0.$$

Applying the closure conditions for the 1-form, we obtain the following necessary conditions for the existence of the entropy function during the process under consideration

$$(32) \quad \partial_F \left(\frac{1}{\theta} \right) = -\partial_e \left[\frac{1}{\mu\theta} \mathbf{t}\mathbf{F}^{-T} \right], \quad \partial_e \left(\frac{1}{\mu\theta^2} \mathbf{q} \cdot \nabla \theta - \frac{1}{\mu} \nabla \cdot \mathbf{k} \right) = \frac{\partial}{\partial t} \left(\frac{1}{\theta} \right),$$

$$(33) \quad \partial_t \left[\frac{1}{\mu\theta} \mathbf{t} \mathbf{F}^{-T} \right] = - \frac{\partial}{\partial \mathbf{F}} \left(\frac{1}{\mu\theta^2} \mathbf{q} \cdot \nabla \theta - \frac{1}{\mu} \nabla \cdot \mathbf{k} \right),$$

$$\frac{\partial \omega_A}{\partial q^B} = 0, \quad (A = 1, 2, 6, B = 3, 4, 5).$$

If the entropy 1-form in (29) is closed and its coefficients are regular (i.e. they have no singularities), this form is exact and the existence of an upper-potential S satisfying the relation $S(\sigma_t) - S(\sigma_0) \geq s$ is ensured (see [8]). Starting from the entropy 1-form, it is possible to investigate and to introduce an extended thermodynamical phase space in a suitable way (see [32]).

5. Clausius-Duhem inequality analysis and heat equation

In this section, taking into account the model developed in [10] for ferroelastic media, we exploit the Clausius-Duhem inequality in the case that one type (size) of grain ($\alpha = 1$) and one kind of anisotropy ($\beta = 1$) is taken into consideration. Applying a Maugin technique [17] (see also Coleman-Noll procedure in [18]) we derive the entropy flux, the residual inequality and the laws of state, where in a relevant result the contributions in the total stress tensor expressed in terms of internal variables are rewritten in terms of the macroscopic variables represented by the Duhamel-Neumann tensor stress and the non-linear Eulerian strain tensor. Finally, the heat equation in the first and the second form is worked out following Maugin in [17].

We consider the entropy inequality $\mu \dot{S} + \nabla \cdot \mathbf{J}_s \geq 0$, with $\mathbf{J}_s = \frac{\mathbf{q}}{\theta} + \mathbf{k}$. Taking into account the positivity of $\theta > 0$ we have

$$(34) \quad \mu \theta \dot{S} + \nabla \cdot (\theta \mathbf{J}_s) - \mathbf{J}_s \cdot \nabla \theta \geq 0.$$

Using in (34) the free energy expression $\Psi = e - \theta S$, the internal energy balance equation (15) and the entropy flux density, we obtain

$$(35) \quad -\mu(\dot{\Psi} + \dot{\theta} S) + \mathbf{p}_{(i)} + \nabla \cdot (\theta \mathbf{k}) - \mathbf{J}_s \cdot \nabla \theta \geq 0.$$

Taking into account that Ψ is a constitutive function of $(\mathbf{E}, \theta, \nabla \theta, \mathbf{q}, \mathbf{\Gamma})$ we have

$$(36) \quad -\mu \left(\frac{\partial \Psi}{\partial \mathbf{E}} \cdot \dot{\mathbf{E}} + \frac{\partial \Psi}{\partial \theta} \dot{\theta} + \frac{\partial \Psi}{\partial \nabla \theta} \cdot \dot{\nabla} \theta + \frac{\partial \Psi}{\partial \mathbf{q}} \cdot \dot{\mathbf{q}} + \frac{\partial \Psi}{\partial \mathbf{\Gamma}} \cdot \dot{\mathbf{\Gamma}} \right) - \mu \dot{\theta} S +$$

$$+ (\mathbf{T} - \sigma^{TE} - \sigma^{HA}) \cdot \dot{\mathbf{F}} \mathbf{F}^{-1} + \nabla \cdot (\theta \mathbf{k}) - \mathbf{J}_s \cdot \nabla \theta \geq 0.$$

Now, using the following expression for the time rate of nonlinear strain tensor (see [33])

$$(37) \quad \dot{\mathbf{E}} = \mathbf{D} - (\mathbf{E} \mathbf{L} + \mathbf{L}^T \mathbf{E}) = \text{Sym}(\dot{\mathbf{F}} \mathbf{F}^{-1}) - (\mathbf{E} \dot{\mathbf{F}} \mathbf{F}^{-1} + \mathbf{F}^{-T} \dot{\mathbf{F}}^T \mathbf{E}),$$

or in components $\dot{E}_{ij} = D_{ij} - (E_{ik} v_{k,j} + E_{jk} v_{k,i})$, with $\mathbf{D} = \frac{1}{2} (\mathbf{L} + {}^T \mathbf{L})$, we have

$$(38) \quad \frac{\partial \Psi}{\partial \mathbf{E}} \cdot \dot{\mathbf{E}} = \frac{\partial \Psi}{\partial \mathbf{E}} \cdot (\dot{\mathbf{F}} \mathbf{F}^{-1} - \mathbf{E} \dot{\mathbf{F}} \mathbf{F}^{-1} - \mathbf{F}^{-T} \dot{\mathbf{F}}^T \mathbf{E}) =$$

$$= \left\{ \left(\frac{\partial \Psi}{\partial \mathbf{E}} \mathbf{F}^{-T} \right) - \left(\mathbf{E}^T \frac{\partial \Psi}{\partial \mathbf{E}} \mathbf{F}^{-T} \right) - \left[\mathbf{E} \left(\frac{\partial \Psi}{\partial \mathbf{E}} \right)^T \mathbf{F}^{-T} \right] \right\} \cdot \dot{\mathbf{F}},$$

where

$$\mathbf{E}^T = \mathbf{E}, \quad \frac{\partial \Psi}{\partial \mathbf{E}} = \left(\frac{\partial \Psi}{\partial \mathbf{E}} \right)^T$$

and the matrix rule $\mathbf{A} \cdot (\mathbf{B}\mathbf{C}) = \mathbf{B} \cdot (\mathbf{A}\mathbf{C}^T) = \mathbf{C} \cdot (\mathbf{B}^T \mathbf{A})$ has been used.

Substituting equation (38) in (36) we obtain the final form for Clausius Duhem inequality:

$$(39) \quad \left[-\mu \left(\frac{\partial \Psi}{\partial \mathbf{E}} - 2\mathbf{E} \frac{\partial \Psi}{\partial \mathbf{E}} \right) + \left(\mathbf{T} - \sigma^{TE} - \sigma^{HA} \right) \right] \mathbf{F}^{-T} \cdot \dot{\mathbf{F}} - \mu \left(\frac{\partial \Psi}{\partial \theta} + S \right) \dot{\theta} - \mu \frac{\partial \Psi}{\partial \nabla \theta} \cdot \nabla \dot{\theta} - \mu \frac{\partial \Psi}{\partial \mathbf{q}} \cdot \dot{\mathbf{q}} - \mu \frac{\partial \Psi}{\partial \mathbf{\Gamma}} \cdot \dot{\mathbf{\Gamma}} + \nabla \cdot (\theta \mathbf{k}) - \mathbf{J}_s \cdot \nabla \theta \geq 0.$$

As \mathbf{T} , σ^{TE} , σ^{HA} , and S are assumed not to depend on $\dot{\mathbf{F}}, \dot{\theta}, \nabla \dot{\theta}$, while the remaining coefficients in (39) may in general depend on their respective factors and (39) has to remain in one sign for any $\dot{\mathbf{F}}, \dot{\theta}, \nabla \dot{\theta}$, taking into account equation (13), we obtain

$$(40) \quad \mathbf{T} = \mu \frac{\partial \Psi}{\partial \mathbf{E}},$$

$$(41) \quad \sigma^{TE} + \sigma^{HA} = 2\mu \mathbf{E} \frac{\partial \Psi}{\partial \mathbf{E}} = 2\mathbf{E}\mathbf{T},$$

$$(42) \quad \frac{\partial \Psi}{\partial \theta} = -S, \quad \frac{\partial \Psi}{\partial \nabla \theta} = 0.$$

Equations (40)-(42) are the *laws of state*. In (41) as new result the sum of σ^{TE} and σ^{HA} expressed in terms of internal variables is derived in terms of the macroscopic variables represented by the Duhamel-Neumann tensor stress \mathbf{T} and the non-linear Eulerian strain tensor \mathbf{E} . At this point, assuming $\nabla \cdot (\theta \mathbf{k}) = 0$, "it is astute to select" the *extra entropy flux density* as (see [17])

$$(43) \quad \mathbf{k} = 0,$$

so that (39) reduces to the following *residual dissipation inequality*

$$(44) \quad \Phi = -\mu \frac{\partial \Psi}{\partial \mathbf{q}} \cdot \dot{\mathbf{q}} - \mu \frac{\partial \Psi}{\partial \mathbf{\Gamma}} \cdot \dot{\mathbf{\Gamma}} - \mathbf{J}_s \cdot \nabla \theta \geq 0.$$

These results are in agreement with the results obtained in [10] as a consequence of Liu's theorem. Very often Φ is split in two parts ("resulting thus in stronger conditions")

$$(45) \quad \Phi_{intr} = -\mathbf{\Pi}^q \cdot \mathbf{q} - \boldsymbol{\eta} \cdot \dot{\mathbf{\Gamma}} \geq 0, \quad \Phi_{th} = -\mathbf{J}_s \cdot \nabla \theta \geq 0, \quad \text{with } \mathbf{\Pi}^q = \mu \frac{\partial \Psi}{\partial \mathbf{q}}, \text{ and } \boldsymbol{\eta} = \mu \frac{\partial \Psi}{\partial \mathbf{\Gamma}},$$

where, in some sense, we recognize the different qualitative nature of the two classes of dissipative processes. Φ_{intr} and Φ_{th} are the intrinsic and thermal dissipations, respectively. Inequalities (45) govern dissipative processes and are in the standard bilinear form in terms of fluxes and associated forces: $\sum_{\beta} X_{\beta} Y_{\beta} \geq 0$, as used in standard irreversible thermodynamics.

Now, in order to obtain the heat equation, we observe that it is none other than a form of energy balance equation. Indeed, on using the free energy expression $\Psi = e - \theta S$, its time derivative and the laws of state in the energy balance equation or, equivalently, "just comparing" entropy inequality (34) and the residual dissipation inequality (44) we deduce the *first general form of the heat equation*

$$(46) \quad \mu \theta \dot{S} + \nabla \cdot (\theta \mathbf{J}_s) = \Phi_{intr},$$

where "the intrinsic dissipation acts like a body source of heat". Now, using the state law (42)₁ and taking into account that the entropy S is a constitutive function of $(\mathbf{F}, \theta, \mathbf{q}, \mathbf{\Gamma})$, we have

$$(47) \quad \dot{S} = - \left[\frac{\partial^2 \psi}{\partial \mathbf{F} \partial \theta} \cdot \dot{\mathbf{F}} + \frac{\partial^2 \psi}{\partial \theta^2} \dot{\theta} + \frac{\partial^2 \psi}{\partial \mathbf{q} \partial \theta} \cdot \dot{\mathbf{q}} + \frac{\partial^2 \psi}{\partial \mathbf{\Gamma} \partial \theta} \cdot \dot{\mathbf{\Gamma}} \right].$$

Finally, substituting equation (47) in (46) and setting $C = -\mu\theta \frac{\partial^2 \Psi}{\partial \theta^2}$, $\boldsymbol{\tau} = \mu \frac{\partial^2 \Psi}{\partial \theta \partial \mathbf{E}}$, $\mathbf{l} = \mu \frac{\partial^2 \Psi}{\partial \theta \partial \mathbf{q}}$, $\mathbf{m} = \mu \frac{\partial^2 \Psi}{\partial \theta \partial \mathbf{\Gamma}}$, we obtain the *second form of the heat equation* in the following compact form:

$$(48) \quad C\dot{\theta} + \nabla \cdot (\theta \mathbf{J}_s) = \Phi_{te} + \Phi_{tq} + \Phi_{t\Gamma}, \quad \text{where}$$

$$(49) \quad \Phi_{te} = \theta \boldsymbol{\tau} \cdot \dot{\mathbf{E}}, \quad \Phi_{tq} = (\theta \mathbf{l} - \boldsymbol{\Pi}^q) \cdot \dot{\mathbf{q}}, \quad \Phi_{t\Gamma} = (\theta \mathbf{m} - \boldsymbol{\eta}) \cdot \dot{\mathbf{\Gamma}}.$$

Here the terms Φ_{te} , Φ_{tq} and $\Phi_{t\Gamma}$ represent the thermoelastic dissipation (due to the thermal and elastic phenomena), the dissipation due to the thermal phenomena and the dissipation due to the interaction between the thermal and the dislocation phenomena, respectively. The non-negativity of the specific heat C follows from the concavity of Ψ with respect to θ ($\frac{\partial^2 \psi}{\partial \theta^2} \leq 0$).

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